*Acta Mineralogica-Petrographica, Szeged 2008, Vol. **48**, pp. 43-48.*

ACTA
Mineralogica
Petrographica

NORDSTRANDITE – A NEW OCCURRENCE FROM HUNGARY

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ABSTRACT

A new occurrence of nordstrandite was described from NE Hungary (Aggtelek-Rudabánya Mts). This mineral appeared in red clayed matrix of conglomerates accumulated in dolines from Triassic platform limestone. It was investigated by X-ray diffractometry, thermal analyses, SEM-EDS. By X-ray analyses it was possible to separate the nordstrandite from other crystalline modifications of $Al(OH)_3$. Nordstrandite appears as individual or radial crystal aggregates in voids (fissures or cavities). In our case the nordstrandite precipitated from amorphous $Al(OH)_3$ gels, from the alkaline phreatic solutions, infiltrating along the fissures of the clayed matrix of conglomerates. Nordstrandite was most probably formed during Late Miocene-Pleistocene.

Key words: Nordstrandite, Aggtelek-Rudabánya Mountains (NE Hungary), terra rossa, X-ray diffractometry, thermal analyses, SEM-EDS.

INTRODUCTION

A half of century ago Van Nordstrand et al. (1956) synthesized a new form of $Al(OH)_3$, what was later named nordstrandite by Papée et al. (1958). Many natural occurrences have since been reported (Table 1). According to Milton et al. (1975), Chao and Baker (1982) and Dani et al. (2001) these may be classified into five groups of natural occurrences of nordstrandite (Table 1):

1. the most important, as a weathering product in bauxitic profiles influenced by carbonatic rocks,
2. as a vein or fissure-filling mineral in dolomitic oil shale,
3. as an alteration product of dawsonite and alumohydrocalcite,
4. as a late-forming mineral in pegmatitic pockets and miarolitic cavities associated with nepheline syenite and sodalite alkaline rocks,
5. weathering product in bauxite derived from alkalic igneous rocks,
6. as recent products in discharged caustic waste.

WHAT IS NORDSTRANDITE?

There are four naturally occurring polymorphous $Al(OH)_3$: gibbsite (JCPDS-cards: 7-0324, 29-0041, 33-0018), bayerite (JCPDS-card: 20-0011), nordstrandite (JCPDS-card: 24-0006), and doyleite (JCPDS-card: 38-0376). All occur in the nature, in natural conditions most frequently gibbsite.

The structure of all these minerals is build up from octahedral layers of $Al(OH)_6$. Differences are in the ways of stacking the layers of $Al(OH)_3$ octahedra. This determinates the symmetry and size of the unit cell of minerals, causing smaller differences. The general structure of the four modifications of $Al(OH)_3$ is presented in the Figure 1.

HISTORICAL BACKGROUND OF THE HUNGARIAN NORDSTRANDITE OCCURRENCES

Nordstrandite was first described in Hungary by Náray-Szabó and Péter (1967), based on the X-ray investigations from the 1–5 μm fraction of brick clays. According to Viczián (1999) although based on chemical analyses these fractions contain $Al(OH)_3$, but these can not be assigned to nordstrandite and bayerite. The determination of the above mentioned two minerals by X-ray analyses is uncertain, because their reflections coincide/interfier with the peaks of kaolinite, chlorite and plagioclase. Consequently the $Al(OH)_3$ determined by Náray-Szabó and Péter (1967) refer to amorphous phase.

Koch (1985) mentioned the presence of nordstrandite in bauxite samples from Csordakút (Transdanubian Range), also investigated by X-ray diffractometry. Albert and Mátrai (1970), Albert et al. (1973) mentioned bayerite and nordstrandite in brick clays (in 1-5μm fraction), but according to Viczián (1999) these were not prooved with certainty.

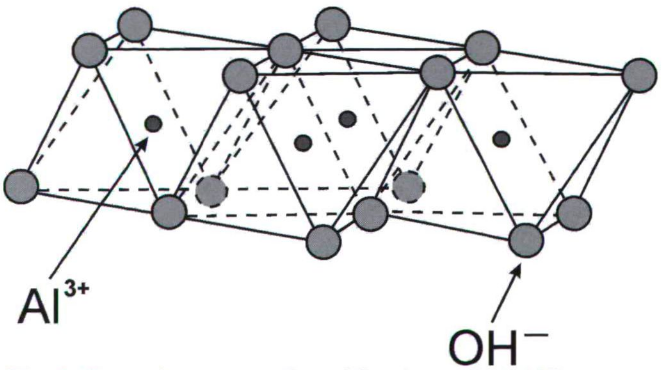

Fig. 1. General structure of modifications of $Al(OH)_3$.

Table 1. Reported geological occurrences of nordstrandite.

Geological setting, location	Type	Associated Al-minerals	Geological age	References
Ferruginous limestone and bauxite (Jamaica)	1	Gibbsite, boehmite	Upper Miocene	Davis & Hill 1973 (in Milton et al. 1975)
Interface limestone and fossil soil derived from basalts and tuffs (Sarawak, Borneo)	1	Gibbsite, diaspore?	Upper Miocene	Wall et al. 1962
Interface limestone and weathered intermediate to basic igneous rock (Guam)	1	Gibbsite, boehmite	Upper Miocene	Hathaway and Schlanger 1962
Interface limestone and weathered volcanic rocks (French Polynesia)	1	Gibbsite, boehmite	Pleistocene	Jamet et al. 1991
Brick clay (Hungary)	1(?)	Gibbsite, bayerite?	Upper Miocene	Náray-Szabó and Péter 1967
Karst limestone, terra rossa (Montenegro)	1	Gibbsite, boehmite	Upper Miocene	Tertian 1966 (in Millot et al. 1975)
Karst limestone, terra rossa (Croatia)	1	Gibbsite, boehmite	Upper Miocene	Marič 1968
Solution cavities on the surface of carbonate rocks (Magnetite mines, Sokolovsko-Sarbay, Russian Federation)	1	Hematite, gibbsite, goethite, magnetite, kaolinite	No data	Kulikova et al. 1974
Karst limestone, terra rossa (Hungary)	1	Kaolinite, hematite, Al-goethite, boehmite	Upper Miocene	This study
Fissure fillings in dolomitic marlstone and oil shale (Rio Blanco, Colorado, USA)	2	Dawsonite	Miocene	Milton et al. 1975
Authigenic in marine and in fluviodeltaic strata (Sydney Basin, New South Wales, Australia)	3	Dawsonite, alumohydrocalcite	Pleistocene	Goldberry and Loughnan 1970
Cavities in nepheline syenite (Narssarssuk, Greenland)	4	No data	No data	Petersen et al. 1976
Pegmatites, miarolitic cavities and xenoliths in nepheline syenite (Mont St-Hilaire, Quebec, Canada)	4	Doyleite, dawsonite	No data	Chao and Baker 1982
Cavities in hauyn-nephelinite (Stradner Hill, Gleichenberg, Austria)	4	Hydrotalcite, motukoreait	Upper Tertiary (Later Pliocene)	Alker et al. 1981
Bauxite derived from phonolite	5	Gibbsite, boehmite	Upper Tertiary (Pliocene?)	Dani et al. 2001
New products in discharged caustic waste (Campbell Island, Southwest Pacific)	6	Bayerite, gibbsite, brucite, pseudoboehmite	Recente	Rodgers et al. 1991

Szakáll and Gatter (1993), Szakáll et al. (2005) consider that the presence of these minerals is not proved properly from the Hungarian occurrences.

The present paper describes the new occurrence of nordstrandite from the Aggtelek-Rudabánya-Mountain, NE-Hungary (Szőlőhegy, Kápolna).

Geological background

The Aggtelek carbonate platform in which the nordstrandite was detected forms a 1–3 km wide belt in the karstified Aggtelek Hills. It strikes NW/SE over a distance of about 7 km between Aggtelek, Jósvafő and Égerszög in NE Hungary. The Triassic formations building up the Aggtelek Karst belong to the Silica Nappe, which forms the uppermost nappe of the Inner West Carpathians (Kozur and Mock 1973).

The Aggtelek platform as part of the non-metamorphosed Aggtelek–Rudabánya Hills belong to the North Pannonian – Inner West Carpathian terrane collage and they are situated in the NE part of the large, composite Pelso Megaunit (Fülöp et al. 1987, Haas ed. 2001) or Pelsonia Composite Terrane (Kovács et al. 2000) (Fig. 2).

On the Middle Triassic carbonate platform lagoonal and reef limestone were deposited. In the Miocene (Sarmatian-Badenian) the Triassic platform carbonates were uplifted and during the denudation period in the karstic doline conglomerates with red clay matrix cement were formed. In the Pleistocene terra rossa with considerable SiO_2 content filled the karstic dolina. The conglomerates were formed during the Miocene or Pleistocene karstic event (Szentpétery et al. 2006).

Nordstrandite occurrences

In the Aggtelek-Rudabánya-Mountains nordstrandite occurs in the karstic doline conglomerates. The clasts of the conglomerate are in red, clayed matrix, sometimes the texture is grain supported. The carbonate clasts are slightly angular and rounded, the diameter of which varies between 2

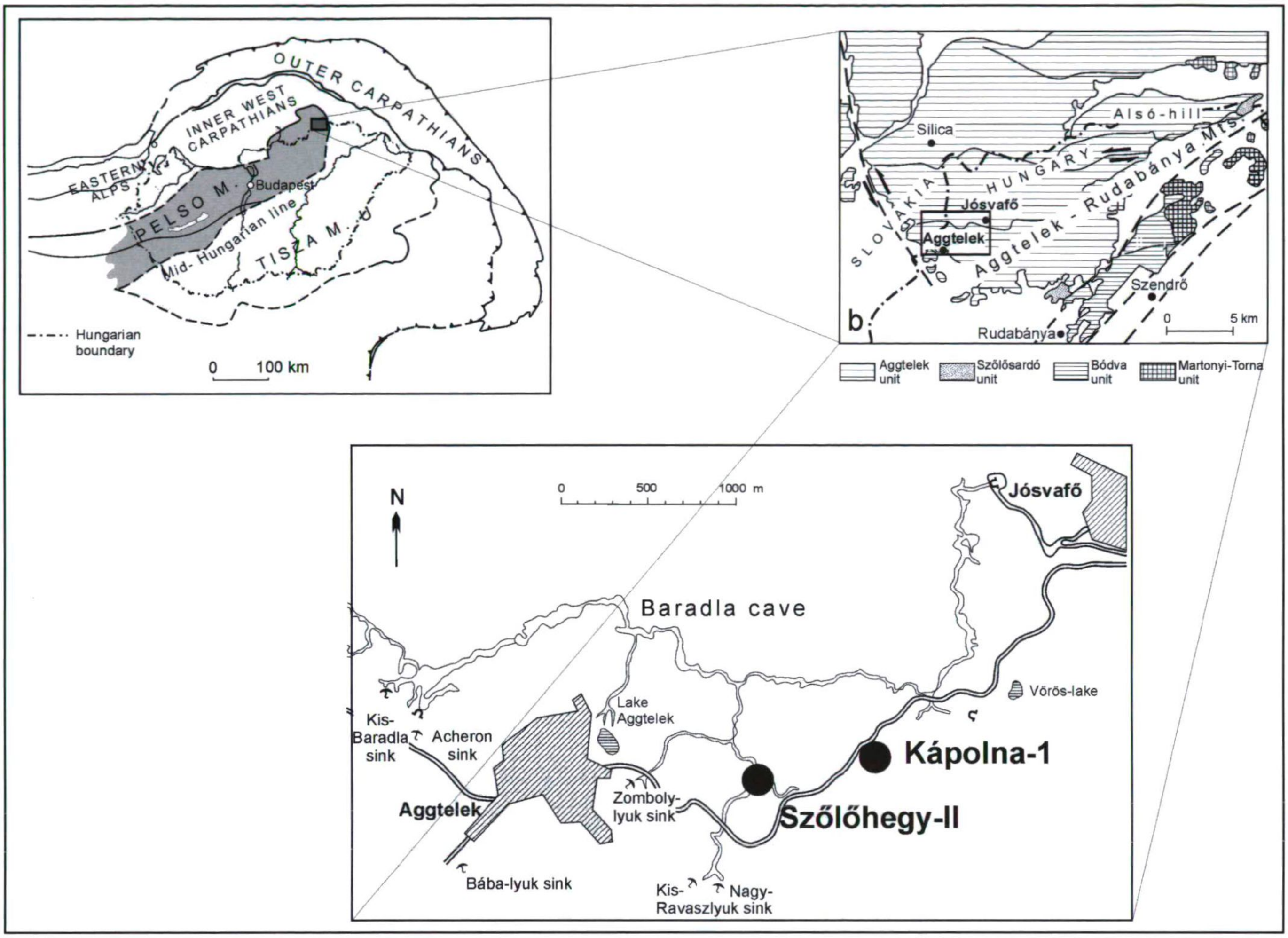

Fig. 2. Sites of nordstrandite occurrences from Aggtelek-Rudabánya-Mountains.

mm and 3 cm. On the edge of the biggest clasts stylolith can be seen, along which dark red clay concentrates. The same dark red clay fills the fractures and fissures, dissecting the matrix of the conglomerate (Fig. 3).

Nordstrandite was first detected by X-ray investigation from the insoluble residue of the red clayed matrix of the carbonate conglomerate deposited in the karstic doline of the Middle Triassic platform limestone. Two samples yielded nordstrandite: Szőlőhegy–II/a (21%) and Kápolna–1 (7%). The present of nordstrandite was confirmed by electron microprobe analyses carried on at Eötvös Loránd University, Department of Petrology and Geochemistry.

METHODS

The X-ray diffraction analyses were done by Philips PW 1710 diffractometer under the following conditions: Cu anti-cathode, 40 kV and 30 mA tube-current, graphite monochromator, goniometer speed 2 °2θ/minute. The mineral composition was calculated on the basis of the relative intensity rates of the reflections characteristic to the minerals, applying the literature or experimental corundum factors on minerals.

The thermoanalytical investigation was executed using a Derivatograph-PC, a computer controlled simultaneous TG, DTG, DTA apparatus. The tests were carried out in ceramic (corundum) crucible, up to 1000 °C. The temperature of the furnace was regulated by a linear heating program at a rate of 10 °C/min. The analytical conditions: air atmosphere, mass of sample about 100 mg, reference material Al_2O_3.

Chemical analysis were preformed on several small crystals by electron microprobe analysis (Department of

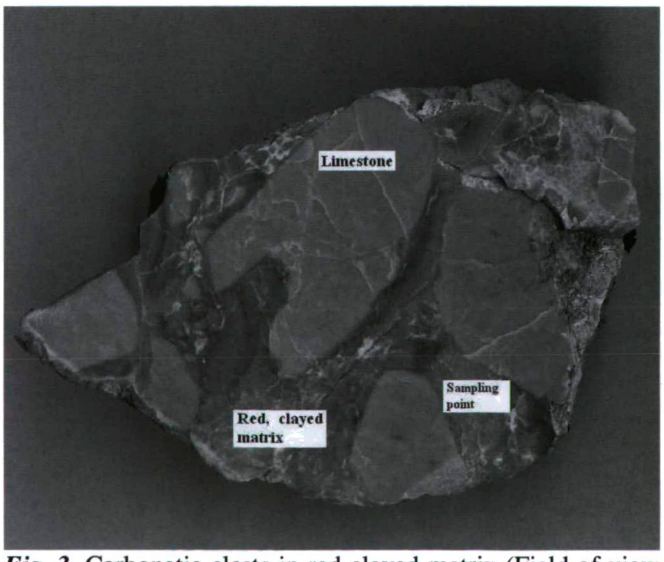

Fig. 3. Carbonatic clasts in red clayed matrix (Field of view is 6 cm wide).

Petrology and Geochemistry of University of Eötvös Lorand), using an AMRAY 1830I/T6 instrument, operated in energy dispersive (EDS) mode (EDAX PV 9800) at 20 kV, with 1-2 nA sample current.

X-RAY DIFFRACTION DATA

Samples of nordstrandite were separated from the conglomerate. The calcite contant was eliminated by dilute acetic acid (20%). The finer clayed matrix was removed from the acid insoluble residues by wet sieving with distilled water. The mineralogical composition (%) of the insoluble residues is presented below (Table 2).

Table 2. The mineralogical composition (%) of the insoluble residues of the nordstrandite-bearing samples.

Minerals	Sample Szőlőhegy-II/a	Sample Kápolna-1
Montmorillonite	2	4
Kaolinite	29	43
Muscovite	13	15
Quartz	5	5
K-feldspars	-	1
Plagioclase	3	3
Dolomite	5	-
Siderite	1	-
Boehmite	1	-
Nordstrandite	21	7
Al-goethite	1	2
Hematite	8	9
Anatase	2	2
Rutile	1	-
Bassanite	2	2
Amorphous	6	7

The X-ray powder diffraction data for the separated insoluble residues are given in Table 3. along with the data for nordstrandite, and other minerals. The X-ray diffractogram of the residual rest in the range of 8 – 26 °2 Θ is shown in Figure 4. Table 4 represents the most important X-ray data for the distinction of different crystalline modifications of $Al(OH)_3$.

On Fig. 5 may be seen the characteristic reflections of nordstrandite (4,790 Å), gibbsite (4,849 Å) and bayerite (4,710 Å) in the investigated sample. Although the 4.794 Å peak of doyleite is situated near the 4.790 Å peak of nordstrandite, but other peaks of the doyleite can not be identified (4.076 Å, 3.096 Å, 2.421 Å, 2.361 Å, 2.325 Å).

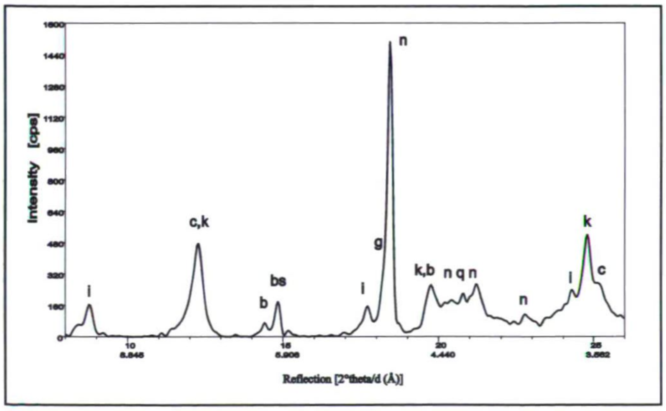

Fig. 4. The X-ray diffractogram of the residual rest in range of 8-26 °2Θ.

Table 3. The X-ray powder diffraction data for the separated insoluble residues.

Szőlőhegy-II/a		Nordstrandite JCPDS 24-0006					Other minerals
d Å	I_{rel}	d Å	I_{rel}	h	k	l	
9.952	10						Muscovite
7.146	31						Kaolinite
6.109	5						Boehmite
5.924	12						Bassanite
4.974	10						Muscovite
4.780	100	4.790	100	0	1	0	
4.467	18						Muscovite. Kaolinite
4.370	12						Kaolinite
4.324	13	4.320	25	0	0	1	
4.250	15	4.210	18	1	0	-1	Quartz
4.193	15						Kaolinite
4.164	18	4.160	12	1	0	0	
3.883	8	3.890	12	1	1	0	
3.841	6						Kaolinite
3.773	7						Kaolinite
3.643	17	3.610	8	1	1	-1	
3.573	36						Kaolinite
3.528	19						Anatase
3.433	7	3.430	6	0	1	1	
3.377	11						Kaolinite
3.340	49						Quartz
3.246	11						Rutile
3.197	11						Plagioclase
3.177	10						Kaolinite
2.965	13						Muscovite
2.884	18						Muscovite. Dolomite
2.797	6						Muscovite
2.699	14						Hematite
2.564	15						Muscovite. Kaolinite
2.514	18						Hematite
2.496	13						Kaolinite
		2.480	12	1	2	0	
2.453	10	2.455	8	1	0	1	
2.388	14	2.393	25	0	2	0	
2.345	16						Kaolinite
		2.333	6	1	2	-1	
2.297	3						Kaolinite
2.283	6						Quartz
		2.271	30	1	1	-2	
2.260	6						Kaolinite
2.202	6						Hematite. Kaolinite
2.189	9						Rutile
2.032	14						Sample holder
2.015	8	2.016	25	1	2	1	
1.994	9						Muscovite. Kaolinite
1.977	7						Muscovite. Kaolinite
		1.945	6	2	2	0	
1.903	5	1.902	20	-2	1	1	
1.783	5	1.784	14	1	2	0	
1.711	5						Muscovite
1.695	7						Muscovite. hematite
1.669	7						Kaolinite
1.600	2	1.598	6	2	3	-1	
1.545	4	1.547	6	3	2	-2	
1.542	5						Quartz
1.503	6						Muscovite
1.490	7						Kaolinite. hematite
1.453	8						Hematite. rutile

Table 4. Crystalline modifications of $Al(OH)_3$

Name	Bayerite	Doyleite	Gibbsite			Nordstrandite
JCPDS Card	20-0011	38-0376	07-0324	29-0041	33-0018	24-0006

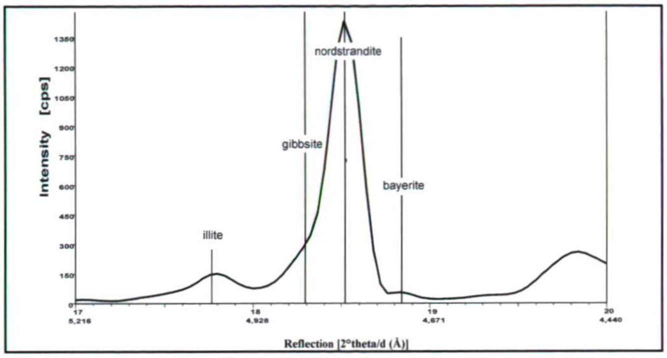

Fig. 5. Characteristic reflections of nordstrandite, gibbsite and bayerite.

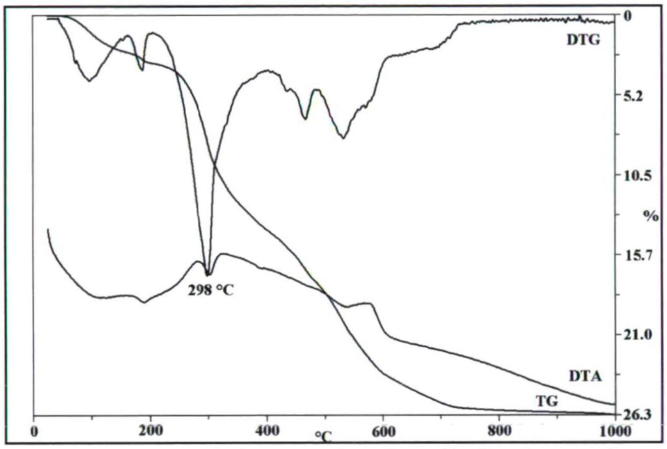

Fig. 6. Thermoanalytical curves of nordstrandite-bearing sample.

THERMAL BEHAVIOUR

Thermal reaction(s) of nordstrandite are very similar to the natural gibbsite (dehydroxylation at about 280 °C). Thermal decomposition appears to proceed as follows:

$$Al(OH)_3 \rightarrow \gamma\text{-}Al_2O_3 + H_2O$$
$$\text{than} \rightarrow \theta\text{-}Al_2O_3 \rightarrow \alpha\text{-}Al_2O_3$$

(Ivanova et al. 1974, Sato 1986, 1987)

Hathaway and Schlanger (1962) mentioned that the D.T.A. of nordstrandite is of limited diagnostic value. Mackenzie (1970) gives this dehydroxylation reaction between 320-330 °C. Van Alker et al. (1956) presented two distinct endothermic reactions, at 110 °C and other stronger at 270 °C. The low temperature water escape at about 100 °C of our sample may be also the reaction of the small amount of bassanite or montmorillonite present in the rock (Fig. 6).

SCANNING-ELECTRONMICROSCOPE AND MICROPROBE

The scanning-electronmicroscope shows the euhedral nordstrandite crystals to be elongated and prismatic (20-40 µm). These appear as individual or radial crystal aggregates in voids (fissures or cavities) in Triassic karst limestone terra rossa (Fig. 7).

GENESIS

The nordstrandite is crystallised at high pH value (7,5-9) (Van Nordstrand et al. 1956). It is not stable on a longer

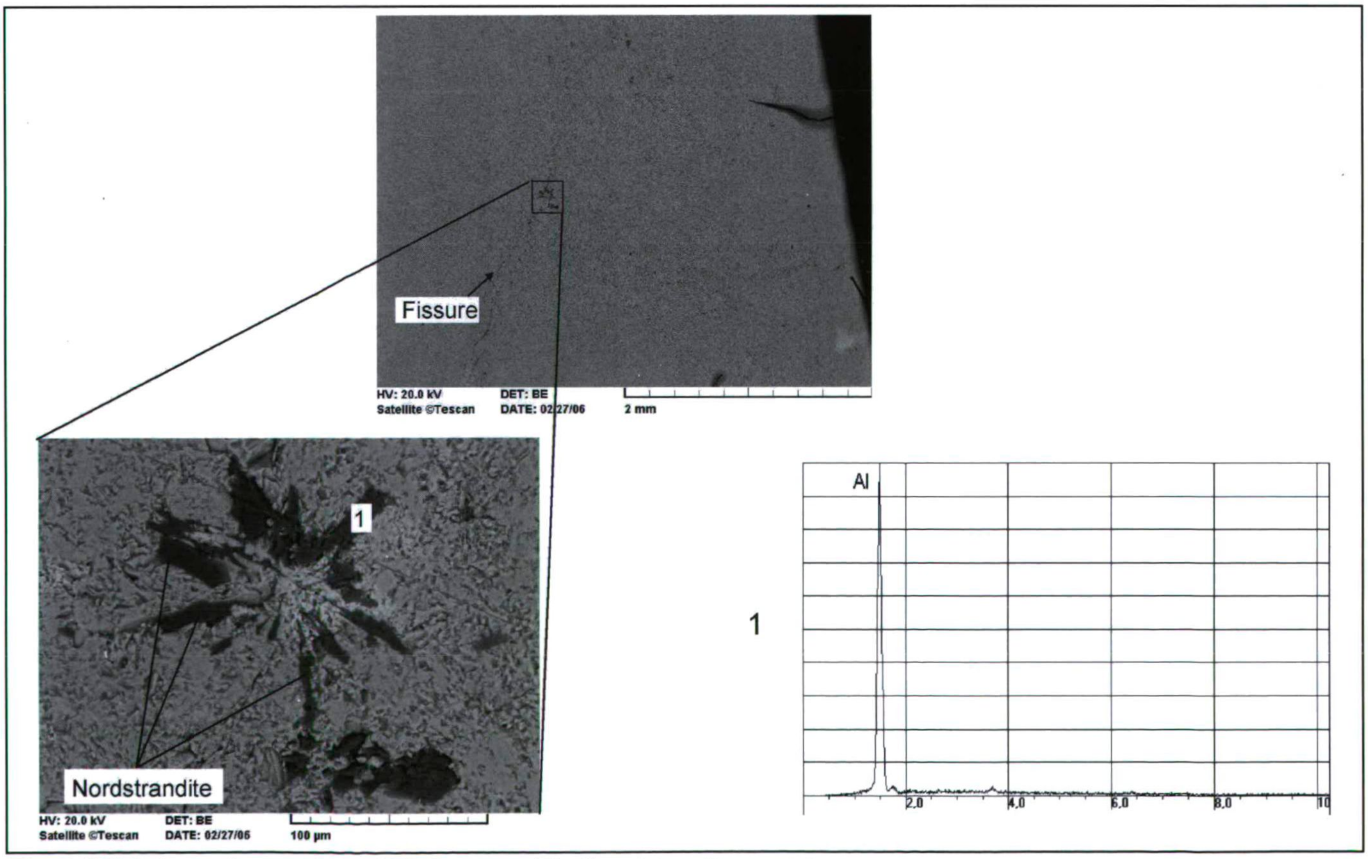

Fig. 7. SEM photomicrographs and EDS spectra (1) of nordstrandite crystals.

time. Its appearance is not likely in deposites older than Miocene, because due to pressure and longer time period it recrysallises into gibbsite. It appears most frequently in karst bauxites, in laterites only by alkaline pH. It can be found frequently in terra rossa deposites formed on carbonatic rocks. In addition to this nordstrandite occurs in cavities of nepheline syenite and in bauxites formed on phonolite (Table 1).

In our case the nordstrandite precipitated from amorphous $Al(OH)_3$ gels, from the alkaline phreatic solutions, infiltrating along the fissures of the clayed matrix of conglomerates accumulated in dolines of Triassic limestones. The huge amount of nordstrandite (sample Szőlőhegy-II/a) refers to in situ precipitation. Taking into consideration the geological history of the area (Szentpétery et al. 2006) nordstrandite was formed during Late Miocene-Pleistocene. Because of many of its properties, in particular the X-ray diffraction pattern, are similar to gibbsite, nordstrandite is probably an unrecognized constituent in many other similar terra rossa occurrences.

ACKNOWLEDGEMENTS

This work was financed by the National Scientific Research Found (OTKA) project number T 037747.

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Received:October 26, 2008; accepted: December 19, 2008